



PCT/GB 2003 / 005198

10/536726

PCT/GB 03/5198



INVESTOR IN PEOPLE

## PRIORITY DOCUMENT

SUBMITTED OR TRANSMITTED IN  
COMPLIANCE WITH RULE 17.1(a) OR (b)

The Patent Office  
Concept House  
Cardiff Road  
Newport  
South Wales  
NP1 8BQ

13 JAN 2004

WIPO

PCT

I, the undersigned, being an officer duly authorised in accordance with Section 74(1) and (4) of the Deregulation and Contracting Out Act 1994, to sign and issue certificates on behalf of the Comptroller-General hereby certify that annexed hereto is a true copy of the international application filed on 02 December 2002 under the Patent Cooperation Treaty at the UK Receiving Office. The application was allocated the number PCT/GB2002/005443.

In accordance with the Patents (Companies Re-registration) Rules 1982, if a company named in this certificate and any accompanying documents has re-registered under the Companies Act 1980 with the same name as that with which it was registered immediately before re-registration save for the substitution as, or the inclusion as, the last part of the name of the words "public limited company" or their equivalents in Welsh, references to the name of the company in this certificate and any accompanying documents shall be treated as references to the name with which it is so re-registered.

In accordance with the rules, the words "public limited company" may be replaced by p.l.c., plc, P.L.C. or PLC.

Re-registration under the Companies Act does not constitute a new legal entity but merely subjects the company to certain additional company law rules.

Signed

Dated

31 DECEMBER 2003

BEST AVAILABLE COPY

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## REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

International Application No. **PCT/GB 2002 / 0 0 5 4 4 3**International Filing Date **02 — 12 — 02**  
**02 DECEMBER 2002**
**United Kingdom Patent Office**  
**PCT International Application**

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference  
(if desired) (12 characters maximum) **15621 MdR**

<b>Box No. I TITLE OF INVENTION</b>	
Catalytic reactor and process	
<b>Box No. II APPLICANT</b> <input type="checkbox"/> This person is also inventor	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)	
Accentus plc 329 Harwell Didcot Oxfordshire OX11 0QJ United Kingdom	Telephone No. <b>01235 43 2037</b> Facsimile No. <b>01235 43 6658</b> Teleprinter No.  Applicant's registration No. with the Office 
State (that is, country) of nationality: GB	State (that is, country) of residence: GB
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input checked="" type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box	
<b>Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)</b>	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)	
BOWE Michael Joseph 17 Balmoral Road New Longton, Preston Lancashire PR4 4JJ United Kingdom	This person is: <input type="checkbox"/> applicant only <input checked="" type="checkbox"/> applicant and inventor <input type="checkbox"/> inventor only (If this check-box is marked, do not fill in below.) Applicant's registration No. with the Office 
State (that is, country) of nationality: GB	State (that is, country) of residence: GB
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input checked="" type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box	
<input checked="" type="checkbox"/> Further applicants and/or (further) inventors are indicated on a continuation sheet.	
<b>Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE</b>	
The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as: <input checked="" type="checkbox"/> agent <input type="checkbox"/> common representative	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)	
MANSFIELD Peter Turquand, LOFTING Marcus John and TALBOT-Ponsonby Clare Josephine all Accentus plc Patents Dept, 329 Harwell Didcot Oxfordshire OX11 0QJ United Kingdom	Telephone No. <b>01235 43 2037</b> Facsimile No. <b>01235 43 6658</b> Teleprinter No.  Agent's registration No. with the Office 
<input type="checkbox"/> Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.	

## Continuation of Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

If none of the following sub-boxes is used, this sheet should not be included in the request.

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

LEE-TUFFNELL Clive Derek  
40 Martin Close  
Poole  
Dorset BH17 7XS  
United Kingdom

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

GB

State (that is, country) of residence:

GB

This person is applicant for the purposes of:

☐ all designated States

☐ all designated States except the United States of America

☒ the United States of America only

☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

MAUDE Jason Andrew  
32 Calderbrook Court  
Meadowbrook Way  
Cheadle, Cheshire SK8 5NX  
United Kingdom

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

GB

State (that is, country) of residence:

GB

This person is applicant for the purposes of:

☐ all designated States

☐ all designated States except the United States of America

☒ the United States of America only

☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

ZIMMERMAN Ian Frederick  
106 Merley Lane  
Wimborne  
Dorset BH21 3AQ  
United Kingdom

This person is:

- ☐ applicant only  
☒ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

GB

State (that is, country) of residence:

GB

This person is applicant for the purposes of:

☐ all designated States

☐ all designated States except the United States of America

☒ the United States of America only

☐ the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below.)

This person is:

- ☐ applicant only  
☐ applicant and inventor  
☐ inventor only (If this check-box is marked, do not fill in below.)

Applicant's registration No. with the Office

State (that is, country) of nationality:

State (that is, country) of residence:

This person is applicant for the purposes of:

☐ all designated States

☐ all designated States except the United States of America

☐ the United States of America only

☐ the States indicated in the Supplemental Box

☐ Further applicants and/or (further) inventors are indicated on another continuation sheet.

## Box No. V DESIGNATION OF STATES

Mark the applicable check-boxes below; at least one must be marked.

The following designations are hereby made under Rule 4.9(a):

## Regional Patent

- ☒ AP ARIPO Patent: GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, MZ Mozambique, SD Sudan, SL Sierra Leone, SZ Swaziland, TZ United Republic of Tanzania, UG Uganda, ZM Zambia, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT (if other kind of protection or treatment desired, specify on dotted line) .....
- ☒ EA Eurasian Patent: AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- ☒ EP European Patent: AT Austria, BE Belgium, BG Bulgaria, CH & LI Switzerland and Liechtenstein, CY Cyprus, CZ Czech Republic, DE Germany, DK Denmark, EE Estonia, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, SK Slovakia, TR Turkey, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- ☒ OA OAPI Patent: BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, GQ Equatorial Guinea, GW Guinea-Bissau, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if other kind of protection or treatment desired, specify on dotted line) .....

## National Patent (if other kind of protection or treatment desired, specify on dotted line):

- |   |  |  |
|---|--|--|
| <input checked="" type="checkbox"/> AE United Arab Emirates .....               | <input checked="" type="checkbox"/> GM Gambia .....                          | <input checked="" type="checkbox"/> NZ New Zealand .....                 |
| <input checked="" type="checkbox"/> AG Antigua and Barbuda .....                | <input checked="" type="checkbox"/> HR Croatia .....                         | <input checked="" type="checkbox"/> OM Oman .....                        |
| <input checked="" type="checkbox"/> AL Albania .....                            | <input checked="" type="checkbox"/> HU Hungary .....                         | <input checked="" type="checkbox"/> PH Philippines .....                 |
| <input checked="" type="checkbox"/> AM Armenia .....                            | <input checked="" type="checkbox"/> ID Indonesia .....                       | <input checked="" type="checkbox"/> PL Poland .....                      |
| <input checked="" type="checkbox"/> AT Austria .....                            | <input checked="" type="checkbox"/> IL Israel .....                          | <input checked="" type="checkbox"/> PT Portugal .....                    |
| <input checked="" type="checkbox"/> AU Australia .....                          | <input checked="" type="checkbox"/> IN India .....                           | <input checked="" type="checkbox"/> RO Romania .....                     |
| <input checked="" type="checkbox"/> AZ Azerbaijan .....                         | <input checked="" type="checkbox"/> IS Iceland .....                         | <input checked="" type="checkbox"/> RU Russian Federation .....          |
| <input checked="" type="checkbox"/> BA Bosnia and Herzegovina .....             | <input checked="" type="checkbox"/> JP Japan .....                           |  |
| <input checked="" type="checkbox"/> BB Barbados .....                           | <input checked="" type="checkbox"/> KE Kenya .....                           | <input checked="" type="checkbox"/> SD Sudan .....                       |
| <input checked="" type="checkbox"/> BG Bulgaria .....                           | <input checked="" type="checkbox"/> KG Kyrgyzstan .....                      | <input checked="" type="checkbox"/> SE Sweden .....                      |
| <input checked="" type="checkbox"/> BR Brazil .....                             | <input checked="" type="checkbox"/> KP Democratic People's Republic .....    | <input checked="" type="checkbox"/> SG Singapore .....                   |
| <input checked="" type="checkbox"/> BY Belarus .....                            | of Korea .....   | <input checked="" type="checkbox"/> SI Slovenia .....                    |
| <input checked="" type="checkbox"/> BZ Belize .....                             | <input checked="" type="checkbox"/> KR Republic of Korea .....               | <input checked="" type="checkbox"/> SK Slovakia .....                    |
| <input checked="" type="checkbox"/> CA Canada .....                             | <input checked="" type="checkbox"/> KZ Kazakhstan .....                      | <input checked="" type="checkbox"/> SL Sierra Leone .....                |
| <input checked="" type="checkbox"/> CH & LI Switzerland and Liechtenstein ..... | <input checked="" type="checkbox"/> LC Saint Lucia .....                     | <input checked="" type="checkbox"/> TJ Tajikistan .....                  |
| <input checked="" type="checkbox"/> CN China .....                              | <input checked="" type="checkbox"/> LK Sri Lanka .....                       | <input checked="" type="checkbox"/> TM Turkmenistan .....                |
| <input checked="" type="checkbox"/> CO Colombia .....                           | <input checked="" type="checkbox"/> LR Liberia .....                         | <input checked="" type="checkbox"/> TN Tunisia .....                     |
| <input checked="" type="checkbox"/> CR Costa Rica .....                         | <input checked="" type="checkbox"/> LS Lesotho .....                         | <input checked="" type="checkbox"/> TR Turkey .....                      |
| <input checked="" type="checkbox"/> CU Cuba .....                               | <input checked="" type="checkbox"/> LT Lithuania .....                       | <input checked="" type="checkbox"/> TT Trinidad and Tobago .....         |
| <input checked="" type="checkbox"/> CZ Czech Republic .....                     | <input checked="" type="checkbox"/> LU Luxembourg .....                      |  |
| <input checked="" type="checkbox"/> DE Germany .....                            | <input checked="" type="checkbox"/> LV Latvia .....                          | <input checked="" type="checkbox"/> TZ United Republic of Tanzania ..... |
| <input checked="" type="checkbox"/> DK Denmark .....                            | <input checked="" type="checkbox"/> MA Morocco .....                         | <input checked="" type="checkbox"/> UA Ukraine .....                     |
| <input checked="" type="checkbox"/> DM Dominica .....                           | <input checked="" type="checkbox"/> MD Republic of Moldova .....             | <input checked="" type="checkbox"/> UG Uganda .....                      |
| <input checked="" type="checkbox"/> DZ Algeria .....                            |  | <input checked="" type="checkbox"/> US United States of America .....    |
| <input checked="" type="checkbox"/> EC Ecuador .....                            | <input checked="" type="checkbox"/> MG Madagascar .....                      |  |
| <input checked="" type="checkbox"/> EE Estonia .....                            | <input checked="" type="checkbox"/> MK The former Yugoslav Republic of ..... | <input checked="" type="checkbox"/> UZ Uzbekistan .....                  |
| <input checked="" type="checkbox"/> ES Spain .....                              | Macedonia .....  | <input checked="" type="checkbox"/> VN Viet Nam .....                    |
| <input checked="" type="checkbox"/> FI Finland .....                            | <input checked="" type="checkbox"/> MN Mongolia .....                        | <input checked="" type="checkbox"/> YU Yugoslavia .....                  |
| <input checked="" type="checkbox"/> GB United Kingdom .....                     | <input checked="" type="checkbox"/> MW Malawi .....                          | <input checked="" type="checkbox"/> ZA South Africa .....                |
| <input checked="" type="checkbox"/> GD Grenada .....                            | <input checked="" type="checkbox"/> MX Mexico .....                          | <input checked="" type="checkbox"/> ZM Zambia .....                      |
| <input checked="" type="checkbox"/> GE Georgia .....                            | <input checked="" type="checkbox"/> MZ Mozambique .....                      | <input checked="" type="checkbox"/> ZW Zimbabwe .....                    |
| <input checked="" type="checkbox"/> GH Ghana .....                              | <input checked="" type="checkbox"/> NO Norway .....                          |  |

Check-boxes below reserved for designating States which have become party to the PCT after issuance of this sheet:

<input type="checkbox"/> .....	<input type="checkbox"/> .....	<input type="checkbox"/> .....
<input type="checkbox"/> .....	<input type="checkbox"/> .....	<input type="checkbox"/> .....

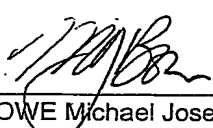
**Precautionary Designation Statement:** In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation (including fees) must reach the receiving Office within the 15-month time limit.)

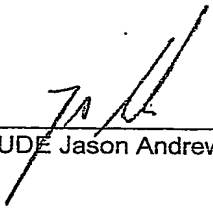
## Supplemental Box

If the Supplemental Box is not used, this sheet should not be included in the request.

1. If, in any of the Boxes, except Boxes Nos. VIII(i) to (v) for which a special continuation box is provided, the space is insufficient to furnish all the information: in such case, write "Continuation of Box No. ...." (indicate the number of the Box) and furnish the information in the same manner as required according to the captions of the Box in which the space was insufficient, in particular:
- (i) if more than two persons are to be indicated as applicants and/or inventors and no "continuation sheet" is available: in such case, write "Continuation of Box No. III" and indicate for each additional person the same type of information as required in Box No. III. The country of the address indicated in this Box is the applicant's State (that is, country) of residence if no State of residence is indicated below;
- (ii) if, in Box No. II or in any of the sub-boxes of Box No. III, the indication "the States indicated in the Supplemental Box" is checked: in such case, write "Continuation of Box No. II" or "Continuation of Box No. III" or "Continuation of Boxes No. II and No. III" (as the case may be), indicate the name of the applicant(s) involved and, next to (each) such name, the State(s) (and/or, where applicable, ARIPO, Eurasian, European or OAPI patent) for the purposes of which the named person is applicant;
- (iii) if, in Box No. II or in any of the sub-boxes of Box No. III, the inventor or the inventor/applicant is not inventor for the purposes of all designated States or for the purposes of the United States of America: in such case, write "Continuation of Box No. II" or "Continuation of Box No. III" or "Continuation of Boxes No. II and No. III" (as the case may be), indicate the name of the inventor(s) and, next to (each) such name, the State(s) (and/or, where applicable, ARIPO, Eurasian, European or OAPI patent) for the purposes of which the named person is inventor;
- (iv) if, in addition to the agent(s) indicated in Box No. IV, there are further agents: in such case, write "Continuation of Box No. IV" and indicate for each further agent the same type of information as required in Box No. IV;
- (v) if, in Box No. V, the name of any State (or OAPI) is accompanied by the indication "patent of addition," or "certificate of addition," or if, in Box No. V, the name of the United States of America is accompanied by an indication "continuation" or "continuation-in-part": in such case, write "Continuation of Box No. V" and the name of each State involved (or OAPI), and after the name of each such State (or OAPI), the number of the parent title or parent application and the date of grant of the parent title or filing of the parent application;
- (vi) if, in Box No. VI, there are more than five earlier applications whose priority is claimed: in such case, write "Continuation of Box No. VI" and indicate for each additional earlier application the same type of information as required in Box No. VI.
2. If, with regard to the precautionary designation statement contained in Box No. V, the applicant wishes to exclude any State(s) from the scope of that statement: in such case, write "Designation(s) excluded from precautionary designation statement" and indicate the name or two-letter code of each State so excluded.

## CONTINUATION OF BOX X

  
 BOWE Michael Joseph

  
 MAUDE Jason Andrew

**Box No. VI PRIORITY CLAIM**

The priority of the following earlier application(s) is hereby claimed:

Filing date of earlier application (day/month/year)	Number of earlier application	Where earlier application is:		
		national application: country or Member of WTO	regional application:* regional Office	international application: receiving Office
item (1) 5 December 2001 (5.12.01)	01 29054.3	GB		
item (2)				
item (3)				
item (4)				
item (5)				

☐ Further priority claims are indicated in the Supplemental Box.

The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) (only if the earlier application was filed with the Office which for the purposes of this international application is the receiving Office) identified above as:

☐ all items    ☒ item (1)    ☐ item (2)    ☐ item (3)    ☐ item (4)    ☐ item (5)    ☐ other, see Supplemental Box

\* Where the earlier application is an ARIPO application, indicate at least one country party to the Paris Convention for the Protection of Industrial Property or one Member of the World Trade Organization for which that earlier application was filed (Rule 4.10(b)(ii)): ....

**Box No. VII INTERNATIONAL SEARCHING AUTHORITY**

Choice of International Searching Authority (ISA) (if two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used):

ISA / EP

Request to use results of earlier search; reference to that search (if an earlier search has been carried out by or requested from the International Searching Authority):

Date (day/month/year)

Number

Country (or regional Office)

**Box No. VIII DECLARATIONS**

The following declarations are contained in Boxes Nos. VIII (i) to (v) (mark the applicable check-boxes below and indicate in the right column the number of each type of declaration):

Number of  
declarations

- |   |  |   |
|---|--|---|
| <input type="checkbox"/> Box No. VIII (i)             | Declaration as to the identity of the inventor   | : |
| <input checked="" type="checkbox"/> Box No. VIII (ii) | Declaration as to the applicant's entitlement, as at the international filing date, to apply for and be granted a patent             | : |
| <input type="checkbox"/> Box No. VIII (iii)           | Declaration as to the applicant's entitlement, as at the international filing date, to claim the priority of the earlier application | : |
| <input checked="" type="checkbox"/> Box No. VIII (iv) | Declaration of inventorship (only for the purposes of the designation of the United States of America)                               | : |
| <input type="checkbox"/> Box No. VIII (v)             | Declaration as to non-prejudicial disclosures or exceptions to lack of novelty   | : |

**Box No. VIII (ii) DECLARATION: ENTITLEMENT TO APPLY FOR AND BE GRANTED A PATENT**

*The declaration must conform to the standardized wording provided for in Section 212; see Notes to Boxes Nos. VIII, VIII (i) to (v) (in general) and the specific Notes to Box No. VIII (ii). If this Box is not used, this sheet should not be included in the request.*

Declaration as to the applicant's entitlement, as at the international filing date, to apply for and be granted a patent (Rules 4.17(ii) and 51bis.1(a)(ii)), in a case where the declaration under Rule 4.17(iv) is not appropriate:

In relation to this international application Accentus plc is entitled to apply for and be granted a patent by virtue of the following:

(ii) Accentus plc is entitled as employer of the inventors:

BOWE Michael Joseph  
LEE-TUFFNELL Clive Derek  
MAUDE Jason Andrew  
ZIMMERMAN Ian Frederick

(ix) This declaration is made for the purposes of all designations (except the designation of the United States of America).

☐ This declaration is continued on the following sheet, "Continuation of Box No. VIII (ii)".

**Box No. VIII (iv) DECLARATION: INVENTORSHIP** (only for the purposes of the designation of the United States of America)  
*The declaration must conform to the following standardized wording provided for in Section 214; see Notes to Boxes Nos. VIII, VIII (i) to (v) (in general) and the specific Notes to Box No. VIII (iv). If this Box is not used, this sheet should not be included in the request.*

**Declaration of inventorship (Rules 4.17(iv) and 51bis.1(a)(iv))  
 for the purposes of the designation of the United States of America:**

I hereby declare that I believe I am the original, first and sole (if only one inventor is listed below) or joint (if more than one inventor is listed below) inventor of the subject matter which is claimed and for which a patent is sought.

This declaration is directed to the international application of which it forms a part (if filing declaration with application).

This declaration is directed to international application No. PCT/..... (if furnishing declaration pursuant to Rule 26ter).

I hereby declare that my residence, mailing address, and citizenship are as stated next to my name.

I hereby state that I have reviewed and understand the contents of the above-identified international application, including the claims of said application. I have identified in the request of said application, in compliance with PCT Rule 4.10, any claim to foreign priority, and I have identified below, under the heading "Prior Applications," by application number, country or Member of the World Trade Organization, day, month and year of filing, any application for a patent or inventor's certificate filed in a country other than the United States of America, including any PCT international application designating at least one country other than the United States of America, having a filing date before that of the application on which foreign priority is claimed.

Prior Applications: .....

I hereby acknowledge the duty to disclose information that is known by me to be material to patentability as defined by 37 C.F.R. § 1.56, including for continuation-in-part applications, material information which became available between the filing date of the prior application and the PCT international filing date of the continuation-in-part application.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name: LEE-TUFFNELL Clive Derek .....

Residence: Poole, Dorset, United Kingdom  
 (city and either US state, if applicable, or country) .....

Mailing Address: c/o Accentus plc, Patents Dept, 329 Harwell  
 Didcot, Oxfordshire OX11 0QJ, United Kingdom .....

Citizenship: British .....

Inventor's Signature: *Clive Tuffnell*  
 (if not contained in the request, or if declaration is corrected or added under Rule 26ter after the filing of the international application. The signature must be that of the inventor, not that of the agent)

Date: 11 November  
 (of signature which is not contained in the request, or of the declaration that is corrected or added under Rule 26ter after the filing of the international application)

Name: ZIMMERMAN Ian Frederick .....

Residence: Wimborne, Dorset, United Kingdom  
 (city and either US state, if applicable, or country) .....

Mailing Address: c/o Accentus plc, Patents Dept, 329 Harwell,  
 Didcot, Oxfordshire OX11 0QJ, United Kingdom .....

Citizenship: British .....

Inventor's Signature: *I. F. Zimmerman*  
 (if not contained in the request, or if declaration is corrected or added under Rule 26ter after the filing of the international application. The signature must be that of the inventor, not that of the agent)

Date: 11 November 2002  
 (of signature which is not contained in the request, or of the declaration that is corrected or added under Rule 26ter after the filing of the international application)

☐ This declaration is continued on the following sheet, "Continuation of Box No. VIII (iv)".



**Box No. VIII (iv) DECLARATION: INVENTORSHIP** (only for the purposes of the designation of the United States of America)  
*The declaration must conform to the following standardized wording provided for in Section 2.14; see Notes to Boxes Nos. VIII, VIII (i) to (v) (in general) and the specific Notes to Box No. VIII (iv). If this Box is not used, this sheet should not be included in the request.*

**Declaration of inventorship (Rules 4.17(iv) and 51bis.1(a)(iv))  
 for the purposes of the designation of the United States of America:**

I hereby declare that I believe I am the original, first and sole (if only one inventor is listed below) or joint (if more than one inventor is listed below) inventor of the subject matter which is claimed and for which a patent is sought.

This declaration is directed to the international application of which it forms a part (if filing declaration with application).

This declaration is directed to international application No. PCT/..... (if furnishing declaration pursuant to Rule 26ter).

I hereby declare that my residence, mailing address, and citizenship are as stated next to my name.

I hereby state that I have reviewed and understand the contents of the above-identified international application, including the claims of said application. I have identified in the request of said application, in compliance with PCT Rule 4.10, any claim to foreign priority, and I have identified below, under the heading "Prior Applications," by application number, country or Member of the World Trade Organization, day, month and year of filing, any application for a patent or inventor's certificate filed in a country other than the United States of America, including any PCT international application designating at least one country other than the United States of America, having a filing date before that of the application on which foreign priority is claimed.

Prior Applications: .....

I hereby acknowledge the duty to disclose information that is known by me to be material to patentability as defined by 37 C.F.R. § 1.56, including for continuation-in-part applications, material information which became available between the filing date of the prior application and the PCT international filing date of the continuation-in-part application.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name: BOWE Michael Joseph

Residence: Preston, Lancashire, United Kingdom  
 (city and either US state, if applicable, or country)

Mailing Address: c/o Accentus plc, Patents Dept, 329 Harwell  
Didcot, Oxfordshire OX11 0QJ, United Kingdom

Citizenship: British

Inventor's Signature: [Signature]  
 (if not contained in the request, or if declaration is corrected or added under Rule 26ter after the filing of the international application. The signature must be that of the inventor, not that of the agent)

Date: 13th November 2002  
 (of signature which is not contained in the request, or of the declaration that is corrected or added under Rule 26ter after the filing of the international application)

Name: MAUDE Jason Andrew

Residence: Cheadle, Cheshire, United Kingdom  
 (city and either US state, if applicable, or country)

Mailing Address: c/o Accentus plc, Patents Dept, 329 Harwell,  
Didcot, Oxfordshire OX11 0QJ, United Kingdom

Citizenship: British

Inventor's Signature: [Signature]  
 (if not contained in the request, or if declaration is corrected or added under Rule 26ter after the filing of the international application. The signature must be that of the inventor, not that of the agent)

Date: 25th November 2002  
 (of signature which is not contained in the request, or of the declaration that is corrected or added under Rule 26ter after the filing of the international application)

☐ This declaration is continued on the following sheet, "Continuation of Box No. VIII (iv)".

## Box No. IX CHECK LIST; LANGUAGE OF FILING

This international application contains:

(a) the following number of sheets in paper form:

request (including declaration sheets) : 9  
 description (excluding sequence listing part) : 16  
 claims : 2  
 abstract : 1  
 drawings : 2

Sub-total number of sheets : 30

sequence listing part of description (actual number of sheets if filed in paper form, whether or not also filed in computer readable form; see (b) below) :

Total number of sheets : 30

(b) sequence listing part of description filed in computer readable form

(i) ☐ only (under Section 801(a)(i))(ii) ☐ in addition to being filed in paper form (under Section 801(a)(ii))

Type and number of carriers (diskette, CD-ROM, CD-R or other) on which the sequence listing part is contained (additional copies to be indicated under item 9(ii), in right column):

This international application is accompanied by the following item(s) (mark the applicable check-boxes below and indicate in right column the number of each item):

Number of items

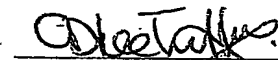
1. ☒ fee calculation sheet : 1
2. ☐ original separate power of attorney : 0
3. ☐ original general power of attorney : 0
4. ☒ copy of general power of attorney; reference number, if any: : 1
5. ☐ statement explaining lack of signature : 0
6. ☐ priority document(s) identified in Box No. VI as item(s): : 0
7. ☐ translation of international application into (language): : 0
8. ☐ separate indications concerning deposited microorganism or other biological material : 0
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  - (iii) ☐ together with relevant statement as to the identity of the copy or copies with the sequence listing part mentioned in left column : 0
10. ☐ other (specify): : 0

Figure of the drawings which should accompany the abstract: 1

Language of filing of the international application: ENGLISH

## Box No. X SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).



LEE-TUFFNELL Clive Derek



ZIMMERMAN Ian Frederick



(On behalf of Accentus plc  
 by virtue of a Power of Attorney  
 dated 5th April 2001)

M. J. LOFTING

For receiving Office use only

1. Date of actual receipt of the purported international application:

02 DECEMBER 2002

/02-12-02

3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:

4. Date of timely receipt of the required corrections under PCT Article 11(2):

5. International Searching Authority (if two or more are competent): ISA /

6. ☐ Transmittal of search copy delayed until search fee is paid

2. Drawings:

☒ received:☐ not received:

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Date of receipt of the record copy by the International Bureau:

Catalytic reactor and process

This invention relates to a chemical process, and to plant including catalytic reactors suitable for use in  
5 performing the process.

A process is described in WO 01/51194 (Accentus plc) in which methane is reacted with steam, to generate carbon monoxide and hydrogen in a first catalytic  
10 reactor; the resulting gas mixture is then used to perform Fisher-Tropsch synthesis in a second catalytic reactor. The overall result is to convert methane to hydrocarbons of higher molecular weight, which are usually liquid under ambient conditions. The two stages  
15 of the process, steam/methane reforming and Fisher-Tropsch synthesis, require different catalysts, and catalytic reactors are described for each stage. The catalytic reactors enable heat to be transferred to or from the reacting gases, respectively, as the reactions  
20 are respectively endothermic and exothermic; the heat required for steam/methane reforming is provided by combustion of methane. A potential problem with this process is that other reactions may occur in the steam/methane reformer reactor, either to generate carbon  
25 dioxide, or to generate coke. It is suggested that the reformer may incorporate a platinum/rhodium catalyst, the reaction being performed at 800°C. The suggested process relies on a steam/methane ratio that is close to 1:1, as the rhodium catalyst is apparently resistant to coking.  
30 An improved way of performing this process has now been found.

According to the present invention there is provided a process for performing steam/methane reforming to  
35 generate carbon monoxide and hydrogen, wherein the gas mixture is caused to flow through a narrow flow channel

between metal sheets separating the flow channel from a source of heat, the flow channel containing a fluid-permeable catalyst structure with a metal substrate to enhance heat transfer, the residence time in the channel  
5 being less than 0.5 second, and both the average temperature along the channel and the exit temperature of the channel being in the range 750°C to 900°C, such that the selectivity in formation of carbon monoxide is above 85%.

10

Preferably the residence time is less than 0.1 s, but preferably at least 0.02 s. It is presumed that such short reaction times enable the process to operate under non-equilibrium conditions, so that only those reactions  
15 that have comparatively rapid kinetics will occur. It is also preferable that the ratio of steam to methane should be in the range 1.2 to 2.0, more preferably 1.3 to 1.6, more preferably about 1.4 or 1.5. Under these conditions the proportion of methane that undergoes reaction can  
20 exceed 90%, and may exceed 95%. Furthermore the selectivity in formation of carbon monoxide rather than carbon dioxide can exceed 90% (selectivity in this context means moles CO/(moles CO + moles CO<sub>2</sub>)).

25 The catalytic reactor preferably comprises a plurality of metal sheets arranged to define first and second flow channels, the channels being arranged alternately to ensure good thermal contact between the fluids in them. Appropriate catalysts should be provided  
30 in each channel, depending on the required reaction. To ensure the required good thermal contact, both the first and the second flow channels are preferably less than 5 mm deep in the direction normal to the sheets. More preferably both the first and the second flow channels  
35 are less than 3 mm deep. Corrugated or dimpled metallic

foils, metal meshes or corrugated or pleated metal felt sheets may be used as the substrate of the catalyst structure within the flow channels to enhance heat transfer and catalyst surface area. Since good heat  
5 transfer is needed for the steam/methane reforming process to achieve high selectivity in the formation of CO, a preferred such structure comprises a metal foil with a thin coating comprising the catalyst material.

10 As described in WO 01/51194, such a reactor may be used for performing methane/steam reforming, the alternate channels containing a methane/air mixture so that the exothermic oxidation reaction provides the necessary heat for the endothermic reforming reaction.  
15 For the oxidation reaction several different catalysts may be used, for example palladium, platinum or copper on a ceramic support; for example copper or platinum on an alumina support stabilised with lanthanum, cerium or barium, or palladium on zirconia, or palladium on a metal  
20 hexaaluminate such as magnesium, calcium, strontium, barium or potassium hexaaluminate, or preferably palladium and platinum on alumina. For the reforming reaction also several different catalysts may be used, for example nickel, platinum, palladium, ruthenium or  
25 rhodium, which may be used on ceramic coatings; the preferred catalyst for the reforming reaction is rhodium or platinum on alumina or stabilised alumina. The catalyst coating is preferably 10 - 50  $\mu\text{m}$  thick. The oxidation reaction may be carried out at substantially  
30 atmospheric pressure, while although the reforming reaction may be carried out at elevated pressure, for example up to 2 MPa (20 atmospheres), operation at atmospheric pressure is preferred, or possibly slightly elevated pressure for example in the range 0 to 200 kPa  
35 above atmospheric pressure.

It will be appreciated that the materials of which the reactor are made are subjected to a severely corrosive atmosphere in use, for example the temperature may be as high as 900°C, although more typically around 5 800°C or 850°C. The reactor may be made of a metal such as an aluminium-bearing ferritic steel, in particular of the type known as Fecralloy (trade mark) which is iron with up to 20% chromium, 0.5 - 12% aluminium, and 0.1 - 3% yttrium. For example it might comprise iron with 15% 10 chromium, 4% aluminium, and 0.3% yttrium. When this metal is heated in air it forms an adherent oxide coating of alumina which protects the alloy against further oxidation; this oxide layer also protects the alloy against corrosion under conditions that prevail within 15 for example a methane oxidation reactor or a steam/methane reforming reactor. Where this metal is used as a catalyst substrate, and is coated with a ceramic layer into which a catalyst material is incorporated, the alumina oxide layer on the metal is believed to bind with 20 the oxide coating, so ensuring the catalytic material adheres to the metal substrate. An alternative structural metal would be Inconel HT800, or another high temperature alloy.

25 For some purposes the catalyst metal might instead be deposited directly onto the adherent oxide coating of the metal (without any ceramic layer).

The gases produced by the steam/methane reforming 30 process described above are preferably then subjected to Fischer-Tropsch synthesis. This may be performed using a second such reactor, with a different catalyst. Where excess hydrogen remains, after the Fisher-Tropsch synthesis, this hydrogen is preferably separated from the 35 desired products, and fed back to the combustion flow channels of the steam/methane reforming reactor.

Combustion of a mixture of methane and hydrogen in the channels has been found to give more uniform temperature, and also enables the combustion reaction to be initiated more readily when the reactor is cold.

5

The invention will now be further and more particularly described, by way of example only, and with reference to the accompanying drawings in which:

10 Figure 1 shows a flow diagram of a chemical process of the invention; and

Figure 2 shows a plan view of a reactor suitable for performing a step of the process shown in figure 1.

15

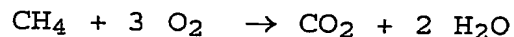
The invention relates to a chemical process for converting methane to longer chain hydrocarbons. The first stage involves steam/methane reforming, that is to say the reaction:

20



This reaction is endothermic, and may be catalysed by rhodium or a platinum/rhodium mixture in a first gas flow channel. The heat required to cause this reaction may be provided by combustion of methane, that is to say:

25

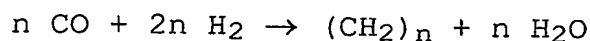


30 which is an exothermic reaction, and may be catalysed by palladium or a platinum/palladium mixture in an adjacent second gas flow channel. Both these reactions may take place at atmospheric pressure, although alternatively the reforming reaction might take place at an elevated pressure. The heat generated by the combustion reaction

35

would be conducted through the metal sheet separating the adjacent channels.

The gas mixture produced by the steam/methane  
5 reforming can then be used to perform a Fischer-Tropsch synthesis to generate a longer chain hydrocarbon, that is to say:



10

which is an exothermic reaction, occurring at an elevated temperature, typically between 200 and 350°C, for example 230°C, and an elevated pressure typically between 2 MPa and 4 MPa, for example 2.5 MPa, in the presence of a  
15 catalyst such as iron, cobalt or fused magnetite, with a promoter such as potassium. The exact nature of the organic compounds formed by the reaction depends on the temperature, the pressure, and the catalyst, as well as the ratio of carbon monoxide to hydrogen. A preferred  
20 catalyst comprises a coating of  $\gamma$ -alumina of specific surface area 140 - 230 m<sup>2</sup>/g with about 10 - 40% (by weight compared to the weight of alumina) of cobalt, and with a ruthenium, platinum or gadolinium promoter, the promoter being .01% to 10% of the weight of the cobalt,  
25 and with a basicity promoter such as ThO<sub>2</sub>. The heat given out by this synthesis reaction may be used to provide at least part of the heat required by the steam/methane reforming reaction, for example a heat transfer fluid such as helium, pressurised water, or Syltherm (trade  
30 mark of Dow Corning) may be used to transfer the heat from a reactor in which the Fischer-Tropsch synthesis is occurring, the heat being used to preheat at least one of the gas streams supplied to the reforming reactor.



Referring now to figure 1, the overall chemical process is shown as a flow diagram. The feed gas 10 consists primarily of methane, with a small percentage (say 10%) of ethane and propane. It is passed through a heat exchanger 11 so it is at about 400°C and is then supplied via a fluidic vortex mixer 12 to a first catalytic reactor 14; in the mixer 12 the feed gas is mixed with a stream of steam that is also at about 400°C, the streams entering the mixer 12 through tangential inlets and following a spiral path to an axial outlet so they become thoroughly mixed. Both streams may be at atmospheric pressure, or for example at a pressure of say 100 kPa above atmospheric. The flows are preferably such that the steam: methane molar ratio (at the steam/methane reforming stage) is between 1.3 and 1.6, preferably 1.4 to 1.5. The first part of the reactor 14 is a pre-reformer 15 with a nickel or platinum/rhodium methanation catalyst at 400°C, in which the higher alkanes react with the steam to form methane (and carbon monoxide); extra steam is required to ensure the desired steam/methane ratio is achieved after this pre-reforming stage (this pre-reformer 15, which may be a separate reactor, would not be required if the feed gas 10 contained substantially no higher alkanes). The second part of the reactor 14 is a reformer 16 with a platinum/ rhodium catalyst, in which the methane and steam react to form carbon monoxide and hydrogen. This reaction may be performed at 850°C.

The heat for the endothermic reactions may be provided by combustion of methane over a palladium or platinum catalyst within adjacent gas flow channels. The catalyst may incorporate  $\gamma$ -alumina as the substrate, coated with a palladium/platinum 3:1 mixture which acts as a catalyst over a wide temperature range, such that the temperature gradually increases from 500°C to 800 or

900°C. The methane/oxygen mixture may be supplied in stages along the reactor 14, to ensure combustion occurs throughout its length.

- 5        The hot mixture of carbon monoxide and hydrogen emerging from the reformer 16 is then quenched by passing through a heat exchanger 18 to provide the hot steam supplied to the vortex mixer 12, and then through the heat exchanger 11 in which it loses heat to the feed gas.
- 10      The mixture is then further cooled to about 100°C by passing through a heat exchanger 20 cooled by water. The gases are then compressed through a compressor 22 to a pressure of 2.5 MPa (25 atm.).
- 15        The stream of high pressure carbon monoxide and hydrogen is then supplied to a catalytic reactor 26 in which the gases react, undergoing Fischer-Tropsch synthesis to form a paraffin or similar compound. This reaction is exothermic, preferably taking place at about
- 20      230°C, and the heat generated may be used to preheat the steam supplied to the heat exchanger 18, using a heat exchange fluid circulated between heat exchange channels in the reactor 26 and a steam generator 28. During this synthesis the volume of the gases decreases. The
- 25      resulting gases are then passed into a condenser 30 in which they exchange heat with water initially at 25°C. The higher alkanes (say C5 and above) condense as a liquid, as does the water, this mixture of liquids being passed to a gravity separator 31; the separated higher
- 30      alkanes can then be removed as the desired product, while the water is returned via the heat exchangers 28 and 18 to the mixer 12.
- Any lower alkanes or methane, and remaining
- 35      hydrogen, pass through the condenser 30 and are supplied to a refrigerated condenser 32 in which they are cooled

to about 5°C. The gases that remain, consisting primarily of hydrogen with carbon dioxide, methane and ethane, are passed through a pressure-reducing turbine 33 and fed via a duct 34 into a storage vessel 35, and hence  
5 through a valve 36 into the combustion channel of the first catalytic reactor 14. The condensed vapours, consisting primarily of propane, butane and water, are passed to a gravity separator 37, from which the water is combined with the recycled water from the separator 31,  
10 while the alkanes are recycled to the feed line 10 so as to be fed into the pre-reformer 15. As indicated by the broken line, electricity generated by the turbine 33 may be used to help drive the compressor 22.

15 When used in this fashion the overall result of the processes is that methane is converted to higher molecular weight hydrocarbons which are typically liquids at ambient temperatures. The processes may be used at an oil or gas well to convert methane gas into a liquid  
20 hydrocarbon which is easier to transport.

From the steam/methane reforming reaction given above one would expect that the appropriate mole ratio for steam to methane would be 1:1. However, at that  
25 ratio there is a significant risk of coking, and a risk that a significant proportion of the methane will not undergo the reaction. Increasing the proportion of steam increases the proportion of methane that reacts, and decreases the risk of coking, although if the proportion  
30 of steam is too high then there is an increased likelihood of carbon dioxide formation. Such excess steam is wasteful of energy and dilutes the reactant gases. It has been found that operating with a steam/methane ratio of between 1.3 and 1.6, preferably  
35 1.4 or 1.5, combined with short residence times that are preferably no more than 100 ms, gives both high

selectivity for carbon monoxide formation and also a very high proportion of methane undergoing reaction. The flow rates through the reformer 16 are preferably such that the residence time is in the range 20 to 100 ms, more preferably about 50 ms.

Such a short residence time enables the reactor 16 to operate under what appears to be a non-equilibrium condition. The competing reaction between carbon monoxide and steam to form the unwanted products carbon dioxide and hydrogen has slower kinetics than the steam/methane reforming reaction to form carbon monoxide and hydrogen; and in the reforming reaction the reverse process has slower kinetics than the forward reaction. The short residence time allows insufficient time for the slower reactions to reach equilibrium. Under these circumstances the proportion of methane undergoing reaction may exceed 90%, and the selectivity for carbon monoxide can exceed 90%.

Experimental measurements of this steam/methane reforming stage are shown in the Table. In each case the steam:methane ratio is 1.5, the gas mixture is preheated to 450° or 550°C and the residence time in the reformer is 50 ms. The reformer has three layers with fifteen channels in all for the steam methane reforming reaction, each channel of breadth 10 mm and length 100 mm, and of depth 1.5 mm, containing a corrugated foil of the same size with a thin alumina coating containing platinum/rhodium catalyst. These channels alternate with four layers of combustion channels to achieve the required heat input.

The temperature was measured at four positions along the length of the centre channel (at 10%, 40%, 60% and 90% of its length), the first and last of these

measurements being tabulated as the inlet and exit temperatures, and the mean of all four temperatures is also tabulated.

Inlet Temp./°C	Exit Temp./°C	Mean Temp./°C	CO Selectivity /%	CH <sub>4</sub> Conversion /%
656	736	712	80.1	80.7
717	851	805	88.6	91.3
727	869	810	90.3	93.7
726	853	810	92.5	93.8
731	866	815	93.0	94.2

5

It will be observed that where both the mean temperature and the exit temperature are above 750°C, the CO selectivity is above 85%. Better selectivity for CO is obtained where the exit temperature is above say  
 10 830°C, preferably above 850°C. The performance of the reactor can also be improved by a pre-treatment, raising the reactor temperature to about 850°C in the presence of hydrogen, as this can improve subsequent catalyst activity.

15

As indicated above, the ideal hydrogen to carbon monoxide stoichiometric ratio to feed to the Fischer-Tropsch synthesis reactor would be 2 moles hydrogen to 1 mole carbon monoxide. This ratio cannot readily be  
 20 obtained by steam/methane reforming: as discussed above, at a steam/methane ratio of 1.0 the resulting gas mixture has a hydrogen to carbon monoxide ratio 3 to 1, and at the elevated steam/methane ratios that must be adopted to avoid coking the hydrogen to carbon monoxide ratio is  
 25 above 3, and may be as high as 4 to 1. Consequently, after the Fischer-Tropsch synthesis reaction has occurred

there will be an excess of hydrogen that remains. Feeding this gas into the combustion channel of the reactor 14 has been found to give a more uniform temperature distribution, and also enables the combustion reaction to be initiated more readily when the reactor is cold (as catalytic combustion can then occur at a temperature as low as 15 or 20°C). The overall thermal efficiency of the process is improved, the amount of methane fed directly to the combustion channels is decreased, and the emission of carbon dioxide to the environment is also reduced.

Referring now to figure 2 a reactor 40 (suitable for example for steam/methane reforming) comprises a stack of plates 42 each of Fecralloy steel, each hexagonal, of side 250 mm and 3 mm thick. It should be appreciated that the reactor 40 is given only by way of example, and that other catalytic reactor designs may alternatively be used. Grooves 44 of width 20 mm and depth 2.5 mm are machined in each plate, extending across each plate 42 from one side to the opposite side, separated by lands 45 of width 3 mm (for simplicity only four such grooves 44 and three such lands 45 are shown in the figure). A carrier foil 46 of Fecralloy steel 50  $\mu$ m thick coated with a ceramic coating of thickness 10 - 20  $\mu$ m containing a catalyst material, and with corrugations 2.5 mm high, can be slid into each such groove 44 (only one is shown). A stack of such plates 42 is assembled, the orientation of the grooves 44 differing by 60° in successive plates 42, and is covered with a flat top plate of Fecralloy steel; the stack is then diffusion bonded together. The corrugated foils 46 are then inserted. Headers 48 are then attached to the faces of the assembled stack. Thus the gas flow channels are defined by the grooves 44; and the orientation of the plates 42 is such that the flow directions in the plates 42 above and below any one plate

42 are at  $120^\circ$  to each other (these grooves being indicated by broken lines and chain dotted lines respectively).

5       The steam/methane mixture is supplied to the header 48a, and the resulting mixture of hydrogen and carbon monoxide emerges through the header 48d. Methane/air mixture is supplied through the headers 48c and 48e (i.e. the headers on either side of the header 48d), so exhaust  
10 gas from the combustion process emerges through the headers 48b and 48f. Hence the gas flows are at least partially counter-current, so that the hottest region in the combustion channels, which is near the inlet to those channels, is closest to the outlet for the steam/methane  
15 reforming reaction. Alternatively in some circumstances there may be partially co-current flow.

      The headers 48 each comprise a simple rectangular cap sealed around its periphery to the outside of the  
20 stack so as to cover one face of the stack. They may be welded onto the outside of the stack. It will be appreciated that after a period of use, if the catalyst in either or both of the channels has become spent, then the headers 48 may be removed or cut off and the  
25 corresponding catalyst-carrying foils 46 removed and replaced. The headers 48 can then be re-attached.

      It will be understood that the type of ceramic deposited on the corrugated foils 46 in the gas flow  
30 channels may be different in successive plates 42 in the stack, and that the catalyst materials may differ also. For example the ceramic might comprise alumina in one of the gas flow channels, and zirconia in the other gas flow channels. The reactor 40 formed from the plates 42 might  
35 also be suitable for performing Fischer-Tropsch synthesis. Because the plates 42 forming the stack are

bonded together the gas flow channels are gas tight  
(apart from communication with headers 48 at each end), ...  
and the dimensions of the plates 42 and grooves 44 are  
such that pressures in the alternate gas flow channels  
5 may be considerably different. Furthermore the pitch or  
pattern of the corrugated foils 46 may vary along a  
reactor channel 44 to adjust catalytic activity, and  
hence provide for control over the temperatures or  
reaction rates at different points in the reactor 40.  
10 There may be more than one layer of corrugated foil  
arranged as a stack in a channel 44. The corrugated  
foils 46 may also be shaped, for example with  
perforations, to promote mixing of the fluid within the  
channels 44. Furthermore parts of the foils 46 may be  
15 devoid of catalyst.

In a modification to the reactor 40, the foils 46  
are again of Fecralloy material, but the catalyst  
material is deposited directly onto the oxide layer of  
20 the Fecralloy. As another alternative the foils 46 might  
be replaced by a metallic mesh of wires or corrugated or  
pleated metal fibre sheets, so providing a large surface  
area for catalyst. It will be appreciated that the  
reactor 40 may be made of other materials, and for  
25 example a reactor 40 for Fischer-Tropsch synthesis may be  
of titanium or stainless steel.

Particularly where the reactor 40 is to be used for  
Fischer-Tropsch synthesis, the gas flow channels 44 for  
30 that reaction may decrease in width, and possibly also  
depth, along their length, so as to vary the fluid flow  
conditions, and the heat or mass transfer coefficients.  
During the synthesis reaction the gas volume decreases,  
and by appropriate tapering of the channels 44 the gas  
35 velocity may be maintained as the reaction proceeds.  
Furthermore the pitch or pattern of the corrugated foils



46 may vary along a reactor channel 44 to adjust catalytic activity, and hence provide for control over the temperatures or reaction rates at different points in the reactor 40. The reactor may also be of a different shape, for example made of a stack of rectangular plates with grooves to define flow channels, the grooves in successive plates being at different orientations - for example at angles of  $0^\circ$ ,  $+45^\circ$ ,  $0^\circ$ ,  $-45^\circ$  etc to the longitudinal axis of the plates, respectively. Headers along the sides of the stack enable gases to flow from one set of channels to the next.

When a reactor such as the reactor 40 is used for reactions between gases that generate gaseous products then the orientation of the channels is not of concern. However if a product may be a liquid, it may be preferable to arrange the reactor 40 so that the flow paths for this reaction slope downwardly, to ensure that any liquid that is formed will drain out of the channels 44. With a rectangular reactor in which the headers provide communication between successive channels, means may be provided in the headers to enhance separation of entrained liquid droplets from the gas flow, or to condense some of the vapours.

It will be appreciated that the overall process described in relation to Figure 1 may be modified in various ways. For example the short-chain alkanes from the separator 37 might be fed into the combustion channel 14. If a long-chain waxy product is found in the Fischer Tropsch reactor 26 then the condenser 30 might cool the product only to say  $60$  or  $80^\circ\text{C}$ .

It will also be appreciated that although the heat for the steam/methane reforming reaction may be provided by catalytic combustion in adjacent channels (as

described above), as an alternative the combustion may take place in an external burner (such as a laminar flow burner), the very hot exhaust gases at about 900 or 1000°C being passed through the second gas flow channels of the reactor 14 in counter-current to the methane flow; this can enable the reacting gases in the reformer 16 to reach a final temperature of as much as 900°C. In this case it is not essential to provide the foils in the combustion channels with ceramic coating or catalyst, but such foils would nevertheless enhance heat transfer between the second gas flow channel carrying the hot exhaust gas and the reactants in the pre-reformer and reformer channels, by transferring heat to the separating plates 42. In the combustion channels of the catalytic reactor 14, if catalytic combustion is used to generate the heat (as indicated), the combustion catalyst may itself be coated with a thin porous inert ceramic layer, so as to restrict the contact of the gas mixture with the catalyst and so restrict the reaction rate particularly at the start of the channel.

Particularly where hydrogen is unavailable, it may be desirable to provide electrical heating by passing an electric current directly through the plates forming the reactor. This may be used initially to raise the temperature for example of the reforming reactor 14 to say 400°C before supplying gases, to ensure catalytic combustion occurs. Such electrical heating may also be used during operation to adjust the reactor temperature. Electrical heating may also be used in the vicinity of the outlet from the reactor 14 to ensure that a temperature of say 900°C is reached by the gases undergoing the reforming reaction.

Claims

1. A process for performing steam/methane reforming to generate carbon monoxide and hydrogen, wherein the gas  
5 mixture is caused to flow through a narrow flow channel (44) between metal sheets (42) separating the flow channel from a source of heat, the flow channel (44) containing a fluid-permeable catalyst structure (46) with a metal substrate to enhance heat transfer, characterised  
10 by the residence time in the channel being less than 0.5 second, and both the average temperature along the channel and the exit temperature of the channel being in the range 750°C to 900°C, such that the selectivity in formation of carbon monoxide is above 85%.  
15
2. A process as claimed in claim 1 wherein the residence time is less than 0.1 s, but preferably at least 0.02 s.
- 20 3. A process as claimed in claim 1 or claim 2 wherein the reforming reaction is carried out at atmospheric pressure.
4. A process as claimed in any one of the preceding  
25 claims wherein the ratio of steam to methane is in the range 1.2 to 2.0.
5. A process as claimed in claim 4 wherein the said ratio is in the range 1.4 to 1.6.  
30
6. A process as claimed in any one of the preceding claims performed using a catalytic reactor defining several adjacent channels for the steam/methane reforming reaction and for an exothermic reaction, arranged  
35 alternately.

7. A process as claimed in claim 6 wherein the exothermic reaction is combustion of gases including hydrogen.
- 5 8. A process as claimed in claim 7 wherein at least some of the hydrogen is that generated by the steam/methane reforming process.
- 10 9. A process as claimed in claim 7 wherein the hydrogen that undergoes combustion is hydrogen that remains after subjecting the carbon monoxide and hydrogen from the steam/methane reforming process to a subsequent Fischer-Tropsch synthesis.
- 15 10. A plant for performing a process as claimed in any one of the preceding claims.

AbstractCatalytic reactor and process

5 Methane is reacted with steam, to generate carbon  
monoxide and hydrogen in a first catalytic reactor; the  
resulting gas mixture can then be used to perform Fisher-  
Tropsch synthesis in a second catalytic reactor. In  
performing the steam/methane reforming, the gas mixture  
10 is passed through a narrow flow channel containing a  
catalyst structure on a metal substrate, and adjacent to  
a source of heat, in a time less than 0.5 s, so that only  
those reactions that have comparatively rapid kinetics  
will occur. Both the average temperature and the exit  
15 temperature of the channel are in the range 750° to  
900°C. The ratio of steam to methane should preferably  
be 1.4 to 1.6, for example about 1.5. Almost all the  
methane will undergo the reforming reaction, almost  
entirely forming carbon monoxide. After performing  
20 Fischer-Tropsch synthesis, the remaining hydrogen is  
preferably used to provide heat for the reforming  
reaction.

Fig.1.

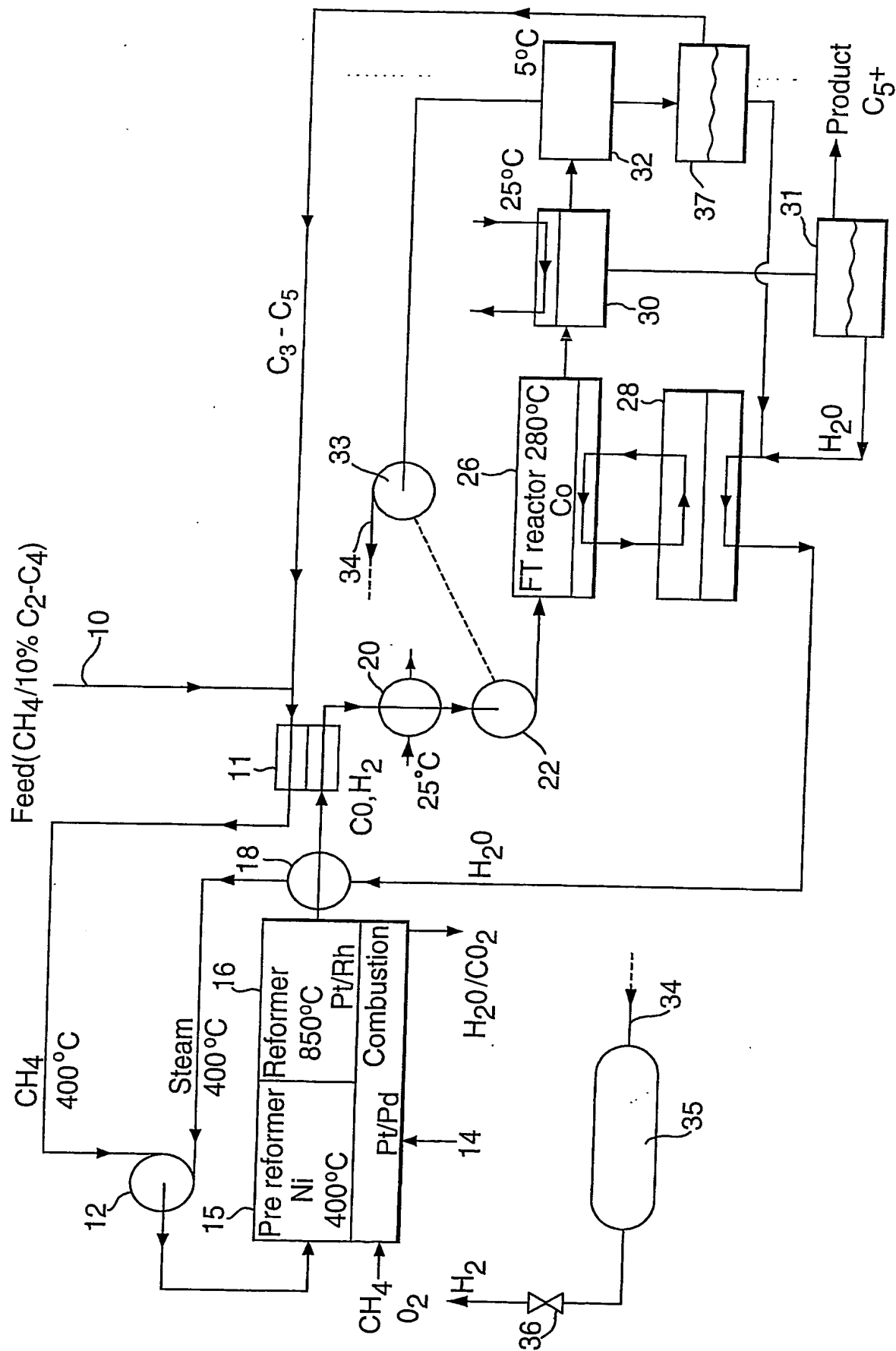
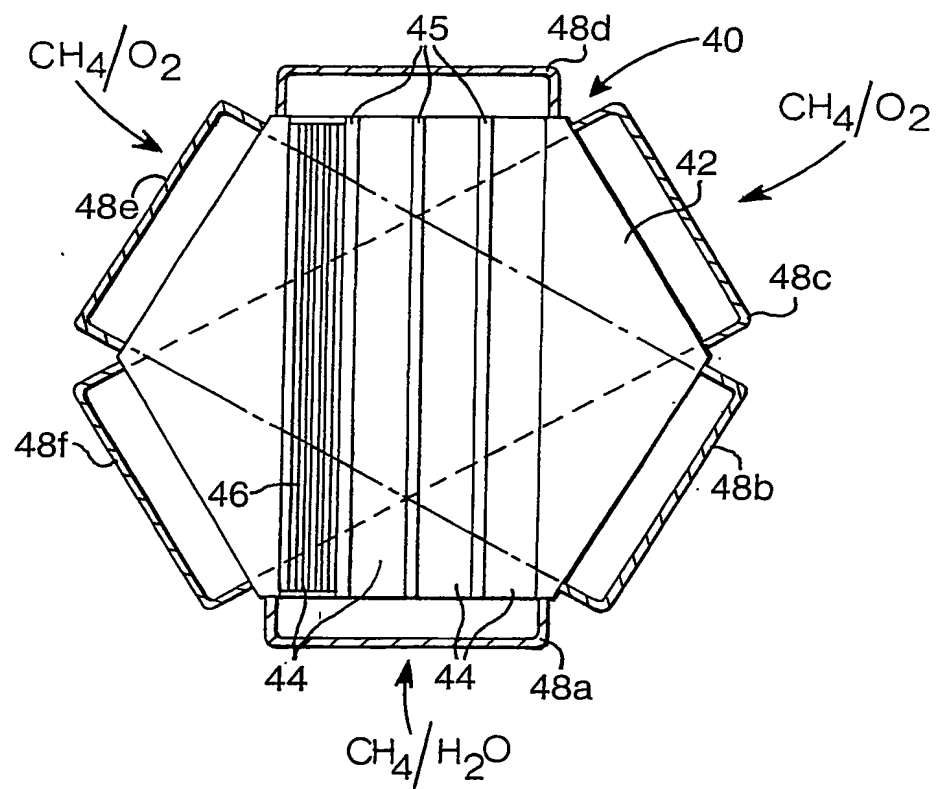


Fig.2.



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